

EXPERIMENTS TO INVESTIGATE CO₂ OCEAN SEQUESTRATION

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KEYWORDS: CO₂; ocean sequestration; environmental impact

ABSTRACT

An international collaboration to investigate the sequestration of CO₂ in the deep ocean was initiated in December 1997. The program is being funded by agencies of the governments of Japan, the U.S., Norway, Canada, and Australia; and Asea Brown Boveri (ABB) of Switzerland. The investigation comprises both experimental and modeling components. During the first phase of the program which runs through March 2002, a field experiment will be conducted in the summer of 2001 in which liquid CO₂ will be injected through various nozzles at a depth of approximately 800 m in the ocean at flow rates ranging from about 0.1 to 1 kg/s. Data will be obtained to develop and validate models that can be applied to predict induced changes to sea water chemistry. This paper describes the planned field experiment and presents selected results from laboratory and modeling studies that have been conducted to support the field experiment.

INTRODUCTION

Anthropogenic emissions of greenhouse gases may precipitate significant changes in global climate. The magnitude and extent of these changes are being actively investigated and debated. Carbon dioxide (CO₂) currently is the most important of these gases due to the preponderant quantities being released into the atmosphere by the combustion of fossil fuels. The latest Intergovernmental Panel on Climate Change (IPCC) assessment indicates that more than 80% of the heat trapping potential of anthropogenic greenhouse gas emissions is associated with CO₂ (UNFCCC, 1997).

International negotiations to establish practices that will stabilize atmospheric concentrations of greenhouse gases have been underway since 1992. In 1997, the Kyoto Protocol established legally-binding greenhouse gas emissions targets for Annex I industrialized countries. For the majority of these countries, the Protocol mandates that a 5% to 8% reduction of CO₂ emissions from 1990 levels be achieved sometime between 2008 and 2012 (UNFCCC, 1998).

Although renewables are being promoted to reduce greenhouse gas emissions, a major transition away from fossil fuels appears unlikely in the near-term. At present, practical and economic factors favor approaches such as increasing the percentage of electricity generated by nuclear power plants, energy conservation, switching from coal and oil to a lower-carbon fuel such as natural gas, and upgrading the efficiencies of fossil fuel energy systems.

Another approach to reduce greenhouse gas emissions into the atmosphere from fossil fuel combustors is to develop techniques to recover, reuse, and/or dispose of the 'fossil' CO₂ released by these devices. One technique that has emerged as a primary candidate for the control of atmospheric carbon emissions involves extraction of CO₂ from flue gases, followed by liquefaction and sequestration underground or in the deep ocean.

Removal and liquefaction of CO₂ from the effluent streams of industrial fossil fuel combustors can be accomplished utilizing existing technologies, albeit at substantial cost (Mori *et al.*, 1993; Herzog *et al.*, 1996). The technical viability of the concept as a means to stabilize emissions therefore depends on whether long term (i.e., of the order of centuries) sequestration of the captured CO₂ from the atmosphere can be achieved. Given the limited range of reuse options, huge quantities of CO₂ will need to be disposed of in an environmentally safe, and cost effective manner. Disposal in the ocean and in subterranean sites such as deep coal seams, spent gas and

oil wells, aquifers, salt domes, and rock caverns has been considered. While geological storage has the potential for very long term sequestration from the atmosphere—of the order of millennia—the world's oceans have a larger capacity to absorb CO₂. It has been estimated that the oceans can accept orders of magnitude more CO₂ than the amount that would be released by oxidation of all known recoverable fossil fuels reserves

The oceans currently remove at least 2 Gigatonnes (i.e., 2×10^{12} kg) of the 6-7 Gigatonnes of carbon released annually by human activity into the atmosphere. Over time, about 80% of this anthropogenic carbon is expected to find its way into the oceans. The slow process of exchange between the surface and deep zones of the ocean, however, constitutes a bottleneck that results in the accumulation of CO₂ in the atmosphere. While direct injection of CO₂ into the depths of the ocean would circumvent this bottleneck, associated impacts on the marine environment in the region surrounding the discharge need to be investigated. CO₂ readily goes into solution in sea water to form dissolved CO₂, and carbonate and bicarbonate ions (Millero & Sohn, 1992). One consequence of this process, however, is a depression of pH. While calcareous materials present in the deep ocean will buffer this effect, and dilution should limit acute changes in sea water chemistry, accurate quantification of these changes is not possible with existing data and models.

Laboratory experiments to investigate CO₂ ocean disposal phenomena have been pursued for over a decade (Masutani & Nihous, 1997). While these experiments have yielded important information, they will never be able to simulate completely the complex, large-scale discharge process. To address this limitation, an R&D program comprising a sequence of three field experiments of increasingly larger scale was authorized via a cooperative agreement signed in December 1997 by representatives of the Governments of Japan, the United States, and Norway. Since its inception, the Governments of Canada and Australia, and Asea Brown Boveri (ABB) of Switzerland, a private sector entity, have joined the program as sponsors.

EXPERIMENTAL PROGRAM

The experimental program has been described previously by Masutani & Nihous (1999), and Masutani (2000). Under the cooperative agreement, three separate field experiments will be conducted in the following sequence: (1) short-term tests of direct release of pure CO₂ at a depth of about 800 m at an open ocean site; (2) longer duration experiments to evaluate acute and chronic biological impacts; and (3) operation of an ocean sequestration test facility over several years to obtain extended-term operational data. Work on the first phase (short-term) field experiment officially was initiated following the signing of the cooperative agreement in December 1997 and will continue through the spring of 2002.

Liquid CO₂ released in the deep ocean is hydrodynamically unstable and will break up into a dispersed droplet phase. Within the depth regime that is accessible using existing pipeline technologies (say, $\leq 1,500$ m), the CO₂ droplets are buoyant and will rise as a plume through the density-stratified water column. A thin, solid hydrate phase may form on the droplet surface that impedes, but does not prevent dissolution (Aya & Yamane, 1992; Teng *et al.*, 1995; Hirai *et al.*, 1995). Sea water will be entrained into the rising droplet plume and transported upward to depths where the ambient water is less dense. Dissolution of CO₂ increases the density of the sea water in the plume. At various points in its ascent, heavy, CO₂-enriched sea water peels away from the plume and subsides to a level of neutral buoyancy (Liro *et al.*, 1992). This CO₂-enriched sea water subsequently is diluted and dispersed by ocean turbulence and currents.

The complex dynamics of the droplet plume in the stratified deep ocean, the effects of hydrates, and dilution and dispersion of CO₂-enriched sea water must be understood in order to address the issues of marine environmental impacts and the effectiveness of sequestration. Models currently are not capable of simulating the behavior of the CO₂ effluent with an acceptable level of confidence, and the fundamental physics of the process are not completely understood.

During the first field experiment, data will be obtained on changes induced in sea water chemistry by the release of pure CO₂. A preliminary sampling of biota and a study of the effects of the discharged CO₂ on naturally occurring bacteria populations also are planned. The specific objectives are:

1. investigate CO₂ droplet plume dynamics through qualitative flow visualization (using mobile video cameras) and quantitative measurements of velocity and pH in the plume and on its margins;
2. clarify the effects of hydrates on droplet dissolution through visualization of the droplet phase and measurements of the vertical extent of droplet rise using scalar indicators (either pH or other added tracers);

3. trace the evolution of CO₂-enriched sea water that peels from the plume by mapping the velocity and relevant scalar (e.g., pH or dissolved inorganic carbon) fields; and
4. assess potential impacts on marine biota by quantifying variations in bacterial biomass, production, and growth efficiency associated with induced changes in seawater pH.

Experiments are scheduled for the summer of the year 2001 at a facility on the west coast of the island of Hawaii. A series of tests with combined duration of about 40 hours will be conducted over two weeks. During these tests, pure liquid CO₂ will be pumped from refrigerated storage tanks installed on a ship to a depth of about 800 m through a small suspended conduit (3 to 6 mm i.d.). The liquid CO₂ will be discharged through different injectors installed at the end of the conduit. The discharge rate will be varied between 0.1 kg/s and 1 kg/s. A range of CO₂ discharge conditions (i.e., mass flow rate; injection velocity; droplet size) will be examined.

Data will be collected employing both fixed and mobile diagnostics. A video system mounted on an ROV (remotely-operated vehicle) will provide flow images of the CO₂ droplet plume. pH sensors and acoustic current profilers will be moored on the sea floor along with the ROV transponders to monitor ambient conditions. Detailed mapping of the scalar and velocity fields will be performed utilizing ROV-mounted instruments that will include conventional salinity, temperature, and pH probes. The ROV will collect data along a three dimensional survey path through the droplet plume and the region of CO₂-enriched sea water generated by the discharge. Water and sediment samples will be collected for chemical and biological analysis and CTD casts will be performed to supplement the data obtained with the moored arrays and ROV.

RESULTS

To support the planning and design of the 2001 field experiment, oceanographic surveys and modeling and laboratory studies are being conducted. A brief overview of this work is provided in this section.

An oceanographic survey of the experimental site off the west coast of the island of Hawaii was conducted during the first week of August 1999. A second survey is scheduled for October 2000. Data has been obtained that: (1) documents the background currents and sea water chemistry and density, and quantifies spatial and temporal variations of these quantities; (2) characterizes ambient bacterial production rates and their response to pH variations; and (3) characterizes the local benthic communities. During the survey, the performance of three methods to measure pH—a conventional glass electrode on the CTD; a novel IS-FET (ion specific field effect transistor) instrument; and shipboard photometric analysis of sea water samples—were compared and evaluated. Accurate and reliable measurements will be critical during the 2001 field experiment, since pH is a primary indicator of the released CO₂. Selected results from the 1999 oceanographic survey have been posted on the project website (<http://www.co2experiment.org>).

Laboratory experiments are being conducted to evaluate injectors and to address concerns related to flow instabilities, flow rate control, and hydrate blockage. Tests are being performed in pressure facilities at the Southwest Research Institute and the University of Hawaii (UH). Figures 1 and 2 present representative video frames obtained in a series of experiments at UH in which liquid CO₂ was pumped into a large pressure vessel to assess the performance of a multiple orifice prototype injector and to investigate hydrate blockage phenomena.

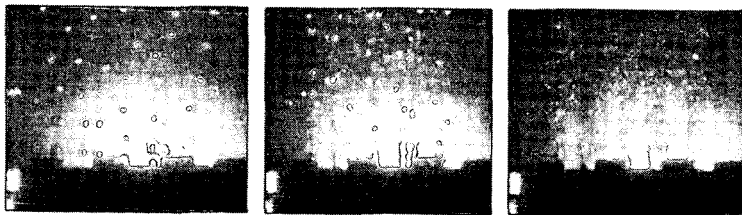


Figure 1 CO₂ injection into a laboratory pressure vessel. The frames show injection at different flow rates through and injector consisting of seven 2 mm diameter ASME sharp-edged orifices; water pressure = 56 bar; water temperature = 2°C.

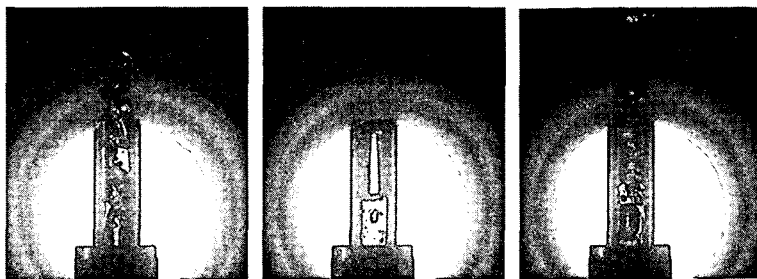


Figure 2 Liquid CO₂ and water flow through a clear nozzle used to investigate hydrate blockage; water pressure = 56 bar; water temperature = 3°C.

Estimates of the height of the droplet plumes and changes in the pH and dissolved CO₂ fields induced by the injection of liquid CO₂ during the experiment are needed to optimize the test plan and instrumentation; and for the Environmental Assessment report and permit applications. Several modeling activities have been pursued by the project team. The most sophisticated supercomputer model produces time-dependent spatial distributions of pH and dissolved and undissolved CO₂ concentrations for user-specified injection scenarios (Alendal *et al.*, 1998). Figure 3 presents one set of results for the Hawaii experiment site when liquid CO₂ is injected at 790 m at a rate of 1 kg/s in a 5 cm/s cross current. It is assumed that the injector produces a monodispersion of 14 mm diameter droplets. The mass transfer coefficient used in these simulations approximates the dissolution of liquid CO₂ droplets covered with a hydrate film. The results imply that the droplet plume will rise less than 150 m before dissolving completely. The lowest pH detected by the simulation for this case was about 6.1 (ambient pH = 7.5).

SUMMARY

A research program to investigate the feasibility of sequestering CO₂ in the deep ocean as a means to reduce the build-up of greenhouse gases in the atmosphere was initiated through an international cooperative agreement signed in December 1997. The program is funded by agencies of the Governments of Japan, the United States, Norway, Canada, and Australia; and Asca Brown Boveri. The program consists of a series of three separate field experiments of increasing magnitude and duration. The first field experiment will be conducted in Hawaii during the summer of 2001. Quantitative data will be obtained on the dissolution and transport of pure CO₂ released at a depth of about 800 m.

To support the planning and design of the field experiment, oceanographic surveys of the site, modeling studies, and laboratory experiments are being pursued. The modeling results suggest that, at the maximum test injection rate of 1 kg/s, a plume comprising relatively large 14 mm diameter droplets will rise less than 150 m before dissolving completely. The lowest pH detected by the simulation for this case was about 6.1 (ambient pH = 7.5). Laboratory studies indicate that a wide range of CO₂ droplet sizes can be generated through appropriate design and operation of the injector. Droplet size is a principal factor that determines the extent of the affected water volume and the magnitude of induced changes to sea water chemistry. Tests also are being performed to assess the potential of hydrate blockage and to devise strategies to avoid this problem.

ACKNOWLEDGMENTS

Support by the Research Institute of Innovative Technology for the Earth (RITE) via subcontracts through the Pacific International Center for High Technology Research and the U.S. Department of Energy under Contract No. DE-FG22-95PC95206 is gratefully acknowledged. This support does not constitute an endorsement by the Department of Energy of the views expressed herein. Experiments at UH were partially supported by the Japan Industrial Technology Association (JITA).

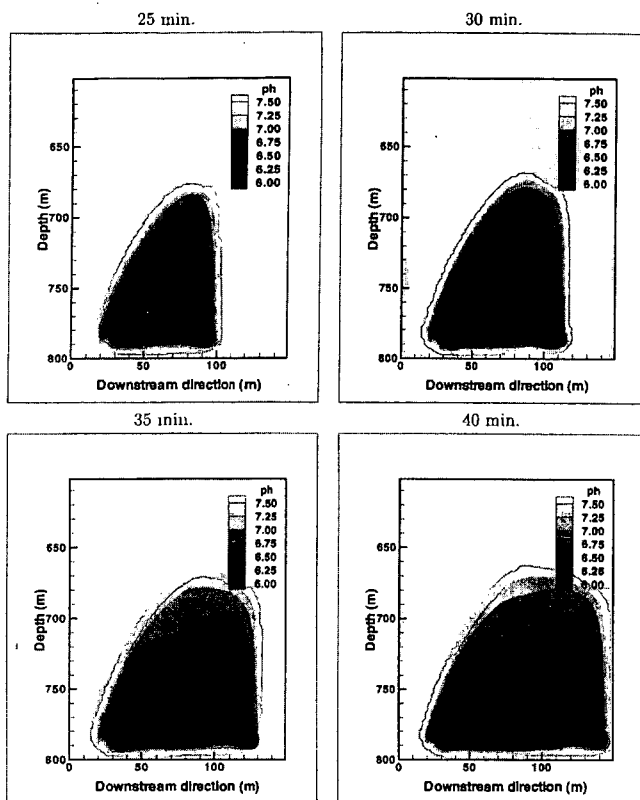


Figure 3 Supercomputer model predictions of pH distributions at different times after the start of injection; ambient pH is 7.5. 14 mm droplets of CO₂ are injected continuously at 1 kg/s at 790 m; mass transfer is assumed to be inhibited by a hydrate film.

REFERENCES

- Alendal G., H. Drange, and F. Thorkildsen, "Two-Phase Modelling of CO₂ Droplet Plumes," Technical Report No.153, NERSC, October 1998.
- Aya, I. And K. Yamane, "Stability of Clathrate-Hydrate of Carbon Dioxide in Highly Pressurized Water," *Proc. Winter Ann. Meeting of ASME*, HTD-Vol. 215, 17, 1992.
- Herzog, H. E. Drake, and E. Adams, "CO₂ Capture, Reuse and Storage Technologies for Mitigating Climate Change," White Paper prepared for U.S. Department of Energy, DE-AF22-96PC01257, 1996.
- Hirai, S., K. Okazaki, N. Araki, Y. Yazawa, H. Ito, and K. Hijikata, "Transport Phenomena of Liquid CO₂ in Pressurized Water Flow with Clathrate Hydrate at the Interface," *Energy Convers. Mgmt* 36(6-8), 471 (1995).
- Liro, C.R., E. Adams, and H. Herzog, 1992. "Modeling the Release of Carbon Dioxide in the Deep Ocean" *Energy Convers. Mgmt* 33(5-8), 667 (1992).
- Masutani, S.M. and G.C. Nihous, "Rig Techniques and Experiments," in *IEA Greenhouse Gas R&D Programme Report on Ocean Storage of CO₂, Workshop 4-Practical and Experimental Approaches* (W. Omerod, ed.), IEA Greenhouse Gas R&D Programme, Cheltenham, UK, 1997.
- Masutani, S.M., "A Field Experiment to Investigate CO₂ Ocean Sequestration," *Proc. Deep Sea and CO₂ 2000*, 2000.
- Masutani, S.M. and G.C. Nihous, "An Update on the International Field Experiment on CO₂ Ocean Sequestration," invited paper, in *Proc. 2nd International Symposium on Ocean Sequestration of Carbon Dioxide*, 1999.
- Millero, F.J. and M.L. Sohn, *Chemical Oceanography*, CRC Press, Boca Raton, FL, 1992.
- Mori, Y., S.M. Masutani, G.C. Nihous, L.A. Vega, and C.M. Kinoshita, "Pre-combustion Removal of Carbon Dioxide from Hydrocarbon-fuelled Power Plants," *Fuel* 72(9), 1293 (1993).
- Teng, H., C.M. Kinoshita, and S.M. Masutani, "Hydrate Formation on the Surface of a CO₂ Droplet in High-Pressure/Low-Temperature Water," *Chemical Engineering Science* 50(4), 559 (1995).
- UNFCCC, Document FCCC/SBI/1997/19/Add.1, from <http://www.unfccc.de/>, 1997.
- UNFCCC, Document FCCC/CP/1997/7/Add.1, from <http://www.unfccc.de/>, 1998.